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R. A. Allen and A. Textoris

RESEARCH NOTE 399
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NATIONAL AERONAUTICS AND SPACE ADMINISTRATION
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ABSTRACT

This work represents a study of the continuum radiation from shock-heated air and nitrogen as a function of density and temperature. For air or nitrogen in which the molecules are completely dissociated yet only partially ionized, radiation produced by the interaction of electrons with neutral atoms will have a stronger density dependence than the radiation produced by electron interaction with positive ions. Intensity measurements of the continuum at 5000 Å were made on the equilibrium region behind incident air and nitrogen shocks in the 9 to 11 mm/μsec range at initial pressures of from 0.1 mm Hg to 1.0 mm Hg. At higher densities, enhanced radiation is observed which can be attributed to the interaction of free electrons with neutral nitrogen atoms. Strong evidence suggests that this radiation is produced by the capture of electrons by nitrogen atoms in the (^{2}D) state. A photoabsorption cross section of 2.6×10^{-16} cm^2 for the N^-(^{1}D) atom at 5000 Å is deduced from the measurements. Besides indicating that a strong N^- continuum exists, these new measurements of Kramers' radiation also indicate that the positive ion radiation contributes less than previously expected and supports the theoretical calculations of Biberman and Norman for the N^+ free-bound radiation.

INTRODUCTION

The present study presents a new interpretation of the radiation from high temperature air or nitrogen which could have important implications for high-speed re-entry or entry into planetary atmospheres. Current estimates of equilibrium radiative heating for super-satellite re-entries or entries have differed by as much as a factor of 10. The source of these discrepancies has been mainly due to various interpretations of N^+ free-bound and free-free Kramers' radiation, which has been regarded as the predominant form of radiation for entry velocities greater than approximately 27,000 ft/sec.

Figure 1 shows a comparison of several generally used calculations for the nitrogen positive ion radiation in air. Total radiation is plotted versus temperature. The various calculations differ by approximately a constant factor, except for optically thick cases. The calculations of Breene,[1] Meyerott,[2] and Lindenmeir[3] essentially agree. For lack of anything better, Meyerott and Lindenmeir have used a theoretical interpretation which assumes that the continuum oscillator strengths of various electron levels in the nitrogen atom are the same as the corresponding electron levels of the hydrogen atom. Biberman and Norman,[4] however, using the quantum defect method of Burgess and Seaton,[5] show that the "hydrogen assumption" regarding the continuum oscillator strengths is not justified for nitrogen. On this basis, the Biberman and Norman calculation is felt to have a better theoretical basis than that of the others. Note, however, that the Biberman and Norman calculation is for free-bound radiation, and a free-free component has been added based on an effective nuclear charge, Z, of 1. The calculations of Kivel and Bailey,[5] are based on a very crude approximation and are the least reliable.

A comparison of the above theoretical predictions with the continuum radiation from a wall stabilized arc has been made by Boldt, who found that the observed radiation exceeded the predictions by a factor of 2. He suggests that the additional radiation is due to radiative capture of electrons by excited N (^{2}D) atoms to form N^-(D). $N^-(^4S)$ cannot exist due to a near zero or negative binding energy for the electron.[15]

The present work represents a study of the continuum radiation from shock-heated air as a function of density. The Biberman and Norman calculation has been shown to be consistent with shock tube measurements performed at low densities and in the infrared.[8] At higher densities, enhanced radiation has been observed. The observed density dependence of the radiation supports Boldt's suggestion and permits the unfolding of the $N^-(^1D)$ continuum portion of the radiation without the necessity of an N^+ radiation calculation.

GENERAL THEORY

The total of the free-bound and free-free radiation at a given wavelength can be described by the Kramers'-Unsöld expression:

$$\frac{dI}{dV\, d\Omega\, d\tilde{\nu}} = \frac{16\pi}{3\sqrt{3}} \frac{e^2}{mc^2} (2\pi\, mkT)^{-1/2} Z_{eff}^{\;2}$$
$$= 1.63 \times 10^{-35}\, T^{-1/2} N_e N_i Z_{eff}^{\;2} \quad \frac{w}{cm^2 ster} \tag{1}$$

where Z_{eff} is the effective charge of the nucleus. N_e and N_i are the particle concentrations of electron and ion, respectively, T is in degrees Kelvin, m is the mass of the electron and c, e, and k are the usual constants. Modifications to this expression have been suggested by Biberman and Norman to account better for the nonhydrogen-like behavior of the free-bound part of the radiation for nonhydrogen ions. However, this modification only effects the Z_{eff} part of the expression, which may or may not be a function of temperature, depending on the atom in question. In any event, at a given wavelength and at constant temperature, the emitted Kramers' radiation will vary as the product $N_e N_i$. Furthermore, for a partially ionized gas at equilibrium, this product is given by the Saha equation:[16]

$$N_e N_i = N_o \frac{g_i g_e}{g_o} \frac{(2\pi\, mkT)^{3/2}}{h^3} e^{-\epsilon_i/kT} \tag{2}$$

where $\frac{g_i g_e}{g_o}$ is the ratio of the internal partition functions of the ion and electron to the internal partition function of the ground state atom; N_o is the number density of neutral atoms; and ϵ_i is the ionization potential.

For normal shocks in air or nitrogen between 9 and 11 mm/μsec, the molecules are completely dissociated, yet only partially ionized; and, therefore, the number density of neutral atoms varies as the gas density. Under these conditions, the N^+ continuum radiation should vary as the first power of the density.

$$\frac{dI}{dV\, d\Omega\, d\tilde{\nu}} \sim N^+ N_e \sim N_o \sim \rho^{1.0} \tag{3}$$

If a neutral atom is involved in the capture or acceleration of an electron in the gas, the N^+ is replaced by the number concentration of the neutral scatterer in the above expression, and

$$\frac{dI}{dV\, d\Omega\, d\tilde{\nu}} \sim N\, N_e \sim \rho^{1.5} . \tag{4}$$

The difference in the density dependence given by Eqs. (3) and (4) is the basis of our method of identifying the radiation due to N^- formation.

EXPERIMENTAL

Shock Tube. The radiation measurements were made on incident air and nitrogen shocks using both a 6-inch and 1.5-inch electric arc-driven shock tube[9] at initial pressures varying from 0.1 to 1.0 mm Hg. Test times ranged from 3 to 7 μsec. Shock speeds were measured by using photomultipliers to observe the radiation from the shock as it passed a series of slits in the shock tube. Generally, the shocks were found to decelerate about 2-4% over the last meter before the test station. The equilibrium temperature and species concentrations behind the shock front were calculated from the initial pressure and shock strength.

Monochromator. Photometric measurements were made in the 5000 to 5300 Å wavelength region using a Jarrel-Ash grating monochromator having a 0.55 mm wide entrance slit. The monochromator was equipped with three 6291 photomultipliers which measured the radiation intensity in three adjacent narrow wavelength bands selected by the monochromator. The middle channel measured the radiation intensity at the wavelength band selected by the monochromator $\pm$ 25 Å, while the red and blue channels measured the radiation intensity at wavelength bands 50 $\pm$ 25 Å above and below that selected by the monochromator, respectively. The wavelength scale of the monochromator was calibrated using a mercury lamp.

In each wavelength region investigated, the detector, monochromator, and shock tube optics were calibrated as a unit, using a standard tungsten ribbon filament lamp calibrated by the National Bureau of Standards. The spectral intensity of the lamp at various wavelengths was calculated using data on the emissivity of tungsten given by DeVos[10] and the transmission of the quartz envelope. In addition, a small secondary lamp which could fit into the shock tube was calibrated against the standard tungsten lamp and used to calibrate the complete optical system, including the shock tube, before each run. This was done to eliminate variations in the transmission of the shock tube wall.

Measurements. Measurements were made in air between 5000 and 5300 Å at three conditions of initial pressure: 0.1, 0.25, and 1.0 mm Hg. The 0.25 and 1.0 mm Hg data were obtained on the 1.5-inch shock tube. However, because of test time limitations, the 0.1 mm Hg data were obtained on a 6-inch shock tube. Both room air and 20% oxygen - 80% nitrogen mixtures were used with no apparent differences.

Figure 2 shows typical radiation profiles of these data for a temperature of approximately 9650°K in the equilibrium region. The level immediately behind the peak at the shock front was taken as the equilibrium level. This peak is seen to decrease relative to the equilibrium level as the pressure increases for constant equilibrium temperature. The overshoot region is also observed to disappear for strong enough shocks at constant initial pressure. This effect is shown in Fig. 3 which shows data at three velocities for an initial pressure of 1.0 mm Hg. For lower initial pressures, the peak is observed to disappear at a higher shock strength. This peak has been observed in the past in strong air[8] and nitrogen[11] shocks and is presumed due to molecular band radiation.

In order to verify that the radiation in the 5000 - 5300 Å region was a continuum, spectra were obtained and a photometric survey was made. The spectrum shown in Fig. 4 was taken with a drum camera spectrograph and shows a homogeneous gas sample at the bottom of the figure which is relatively clear of line radiation in this wavelength region.

The presence of a continuum in this region was also verified by photometric measurements with 50 Å resolution which show very little structure as a function of wavelength. These measurements are shown in Fig. 5 for equilibrium air at $P_1 = 1.0$ mm Hg, $\rho_2/\rho_1 = 15.05$ U_s = 10 mm/μsec, T = 10,500°K. The brackets indicate the standard deviation of the data at each wavelength. Also plotted in the figure are the nitrogen arc measurements of Morris and Boch[12] which were obtained at the same temperature, but at an electron concentration of 3.0×10^{16} per cc as compared with 4.0×10^{16} per cc for the equilibrium conditions of the air shock tube data in the figure. The nitrogen atom concentration of the arc measurement was 6.4×10^{17} per cc as compared with 5.3×10^{17} per cc for the shock tube measurements. The arc data have been scaled to the same product of nitrogen atom and electron concentrations as that of the shock tube data. Measurements made in the infrared as part of a wavelength survey are also plotted in the figure.

The intensity of the 5000 - 5300 Å continuum obtained at the three different initial pressures is plotted in Fig. 6 versus shock velocity. The three verticle lines for the three different densities in the figure to which nearby data points have been extrapolated are for an equilibrium temperature of 9650°K. The data points have been extrapolated along the dotted lines inferred from the velocity dependence of the 1.0 mm Hg data which cover the widest velocity range.

Photometric measurements were also obtained in pure nitrogen in the 5000 - 5300 Å region, and at an initial pressure of 1.0 mm Hg. These data are shown in Fig. 7. The theoretical lines will be discussed in the next section.

ANALYSIS

The photometric measurements at three conditions of density, but at the same wavelength and temperature, are replotted in Fig. 6 versus density. Also plotted in the figure are the arc measurements performed in nitrogen by Morris and Boch.[12] The radiation produced by the capture of electrons by neutral O plotted in this figure is based on the measurements of the photoabsorption cross-section of $O^-(^2P)$ made by Branscomb and Smith.[13] This continuum as well as the calculated $N_2^+(1-)$ radiation[17] has roughly the correct density dependence but falls a factor of 10 below the measurements. As stated before, the Biberman and Norman calculation is felt to be the most reliable calculation for the free-bound positive ion radiation. This calculation plus the assumption that the Z_{eff} for the free-free ion radiation is 1, is used to calculate the theoretical line in the figure for the N^+ and O^+ ion radiation. This theoretical line not only has the wrong density dependence to be compatible with the data but also falls well below the higher density data. From the argument made in the second section regarding the density dependence of Kramers' radiation and also because the measurements are higher than the radiation to be expected from other sources, it is proposed that the remainder of the radiation is that of the $N^-(^1D)$ continuum produced by the capture of an electron by an excited $N(^2D)$ atom as suggested by Boldt.

The photodetachment cross-section, σ, for $N^-(^1D)$ may be inferred using detailed balancing arguments from the free-bound radiation.

$$\frac{dI}{dV\, d\Omega\, d\lambda} = \sigma\, 2h^2\, \lambda^{-5}\, [N^-]\, e^{-hc/kT\lambda} \qquad (5)$$

The photodetachment cross-section can be inferred once the $N^-(^1D)$ concentration is known. $N^-(^1D)$ is metastable and spontaneously breaks up into a $N(^4S)$ atom and a free electron. The decay time for this process is between 10^{-6} and 10^{-7} sec.[7] As long as this radiationless decay time is much longer than the collisional time for the destruction of $N^-(^1D)$ then the Saha equation (2)[16] can be used to calculate the $N^-(^1D)$ concentration. This negative ion has a calculated electron binding energy of 1.1 ev[7, 15] and an internal partition function of 5. In the Saha equation used to calculate $N^-(^1D)$, $N^-(^1D)$ replaces N_o in expression (2) and $N(^2D)$ replaces N_i. $N(^2D)$ has a degeneracy of 10 and lies 2.38 ev above the $N(^4S)$ ground state which has a degeneracy of 4. N_e, the electron concentration has been calculated by applying the Saha equation to the equilibrium gas conditions and assuming the presence of no negative ions. The reason that this can be done is that N^- is much less than N_e. For example, the resultant concentrations calculated for the equilibrium region behind an incident air shock, P_i = 1 mm Hg, U_s = 9.6 mm/μsec are

$$\begin{aligned} T_e &= 9650^\circ K \\ N_e &= 1.5 \times 10^{16}\ cm^{-3} \\ N_i &= 1.3 \times 10^{16}\ cm^{-3} \\ N^-(^1D) &= 5.7 \times 10^{11}\ cm^{-3} \\ N_o &= 8.4 \times 10^{17}\ cm^{-3} \end{aligned}$$

At approximately these same conditions, Boldt[7] has shown that the radiationless decay time for the break-up of $N^-(^1D)$ is longer than its collisional time for destruction by at least a factor of 20 and, consequently, the use of the Saha equation is justified for the calculation of $N^-(^1D)$.

If one assumes the N + e radiation is predominantly free-bound, then the data of Figs. 7 and 8 imply a photoabsorption cross-section for $N^-(^1D)$ of $2.6 \times 10^{-16}\ cm^2$ with approximate error of $\pm$ 30%. Taylor[14] has made measurements in the infrared beyond the 1.1 ev cut-off edge of the $N^-(^1D)$, and inferred an effective Z_{eff}^2 for the free-free scattering of electrons by neutral nitrogen atoms of .009. This finding is consistent with the notion that the predominant N + e radiation at 5000 Å is free-bound.

The 1.0 mm Hg air data of Fig. 5 are replotted in Fig. 9 for comparison with the theory which assumes an $N^-(^1D)$ continuum based on a $\sigma_{N^-(^1D)}$ of $2.6 \times 10^{-16}\ cm^2$. Other theory lines in the figure are the contributions to be expected from O^+ and N^+ ion radiation according to Biberman and Norman and the O_{fb} according to Branscomb and Smith. The total theory line based on these assumptions is within the scatter of the points, although slightly low, and has the expected velocity dependence.

Additional evidence for this interpretation of the radiation is the nitrogen arc data recently obtained by Morris and Boch.[12] These data, as Boldt's, were obtained at atmospheric pressure. A portion of the data taken at 5000 Å is plotted versus temperature in Fig. 6. Theoretical calculations are placed on this figure which are based on the present hypothesis regarding the $N^-(^1D)$ continuum. The Biberman and Norman calculation is again used for the free-bound N^+ radiation. The Z_{eff}^2 used for the free-free component of the N^+ radiation is taken to be 1.

DISCUSSION

In this study, the assumption is made that $N^-(^1D)$ does exist and that its electron binding energy is approximately 1.1 ev. There has been no direct experimental verification of the $N^-(^1D)$ ion and the main evidence for its existence has been circumstantial. The difficulty of obtaining direct measurements is not unexpected due to the short lifetime (10^{-6} - 10^{-7} sec) of this metastable ion.[7] Boldt used a binding energy of 1.1 ev for this ion and this value has been adopted for the purposes of our analysis. There is another value quoted in the literature of 1.05 ev obtained by Edlén[15] using an isoelectric extrapolation method. Edlén also uses this method to calculate electron affinities for other negative ions and obtains agreement to within 10% of the measured values for C^-, O^-, F^-, S^- and Cl^-.

Another assumption made in this study is that local thermodynamic equilibrium exists in the region behind the relaxation region at the shock front. Implicit in this assumption is that collisional destruction of N^- is the dominant mechanism over destruction by its radiationless decay. As pointed out before, Boldt[7] has shown that this condition was fulfilled for his arc measurements which corresponded to our higher density measurements. This assumption regarding thermodynamic equilibrium is also the basis on which the temperatures and species concentrations are calculated. Although there is large scatter inherent in the shock tube measurements, agreement between them and arc measurements is gratifying. This argument strongly indicates that the gas conditions are properly calculated for both experiments.

The present interpretation regarding Kramers' radiation as arrived at by the shock tube work and existing arc data, is that the Biberman and

Norman calculations are essentially correct for the N^+ free-bound radiation, and that there exists a strong $N^-(^1D)$ continuum which will predominate at increased densities. This $N^-(^1D)$ continuum will have a cut-off in the 1.1 to 1.2 μ wavelength region, corresponding to the binding energy.

To point up the temperature and density regimes where the $N^-(^1D)$ continuum would be important for equilibrium radiation, a graph of intensity at 5000 Å for various densities is shown in Fig. 7. The N continuum is based on $\sigma_{N^-(^1D)}$ of 2.6×10^{-16} cm^2 at 5000 Å; and the N^+ continuum, on the calculations of Biberman and Norman.

Before one can make a precise calculation of the radiative heating from this source, it is necessary to know the wavelength dependence for the photoabsorption cross-section for the $N^-(^1D)$ ion. Generally speaking, however, re-entries which decelerate at low altitudes will encounter more radiative heating than previously expected, while re-entries which decelerate at high altitudes will encounter equilibrium radiation heating from Kramers' radiation which is less than that predicted by Kivel and Bailey, Meyerott, and Breene.

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Credit is to be given to J. C. Keck for his helpful suggestions and to J. Carlson and J. McEachern who ably assisted in the experiment. Special thanks is to be given to J. Morris and G. Boch for many fruitful discussions regarding the arc measurements.

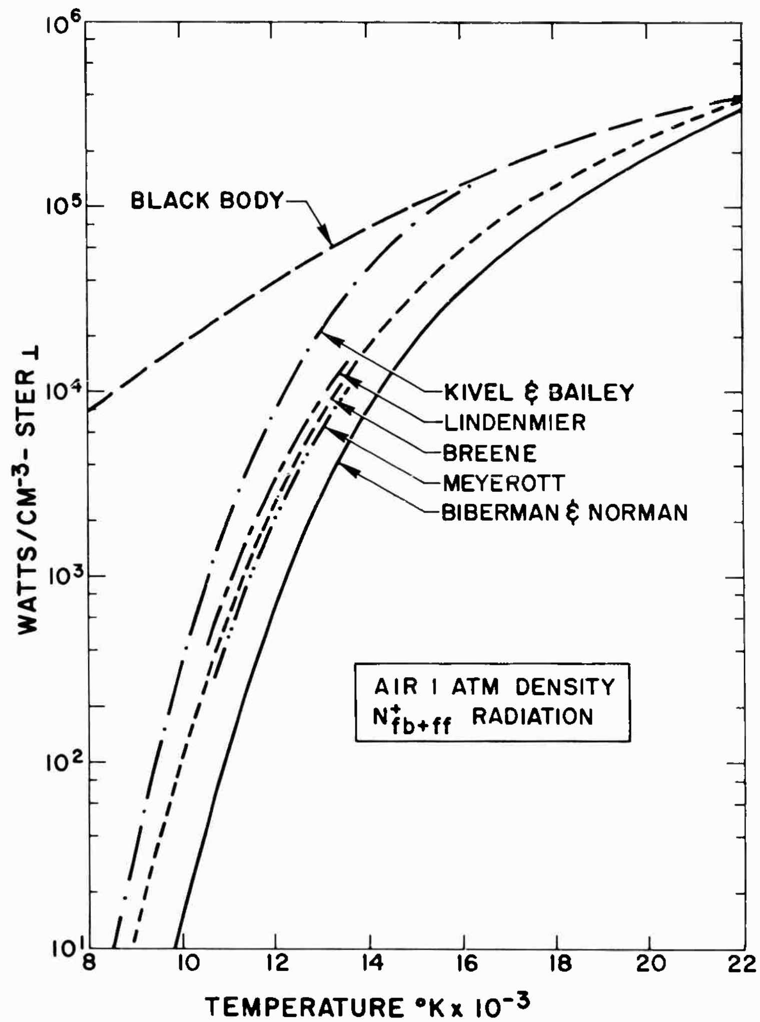

Fig. 1 Comparison of existing calculations for the N^{+} ion Kramers' radiation at 1 atm density showing the large factor by which they differ. The discrepancy between these various calculations is discussed in the text.

A 1367

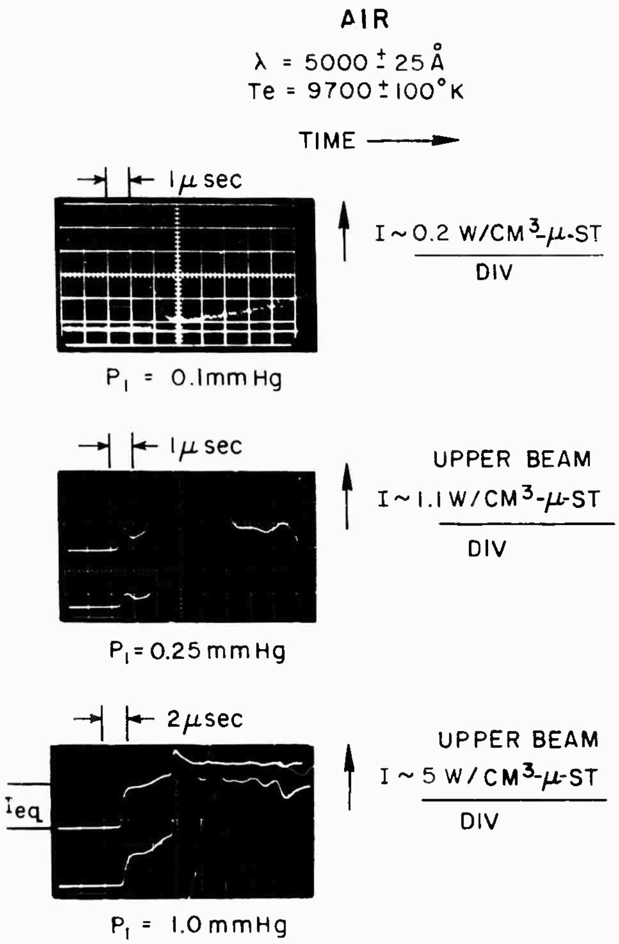

Fig. 2 Typical oscillograms of radiation profiles of shock-heated air in 5000 to 5300°K wavelength region at a P_1 of 0. 1, 0. 25 and 1. 0 mm Hg. The equilibrium level is taken to be that immediately behind the peak at the shock front.

A1605

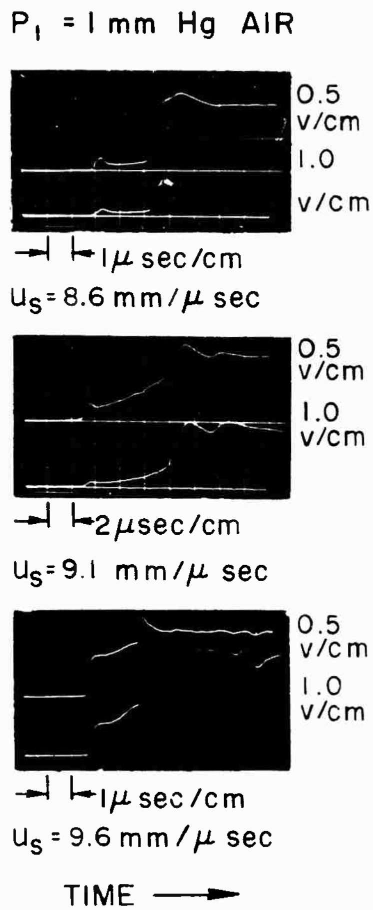

Fig. 3 Oscillograms of radiation profiles in the 5000-5600°K wavelength region showing how the peak in the radiation at a given pressure disappears for strong enough shocks.

A1432

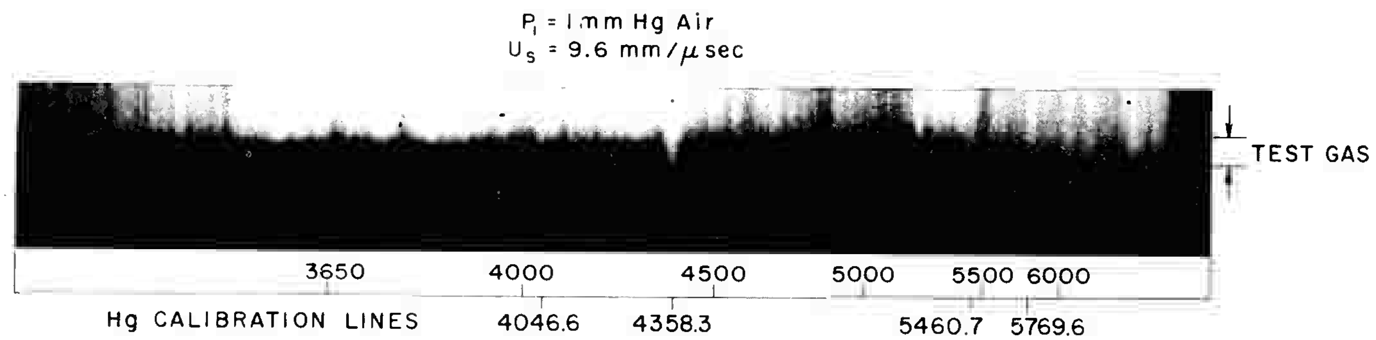

Fig. 4 Drum camera spectrum of an incident air shock, $P_1 = 1$ mm Hg, showing the 5000-6000 Å region to be relatively clear of line radiation. The homogeneous gas sample is at the bottom of the figure with the luminous driver gas following.

A1437

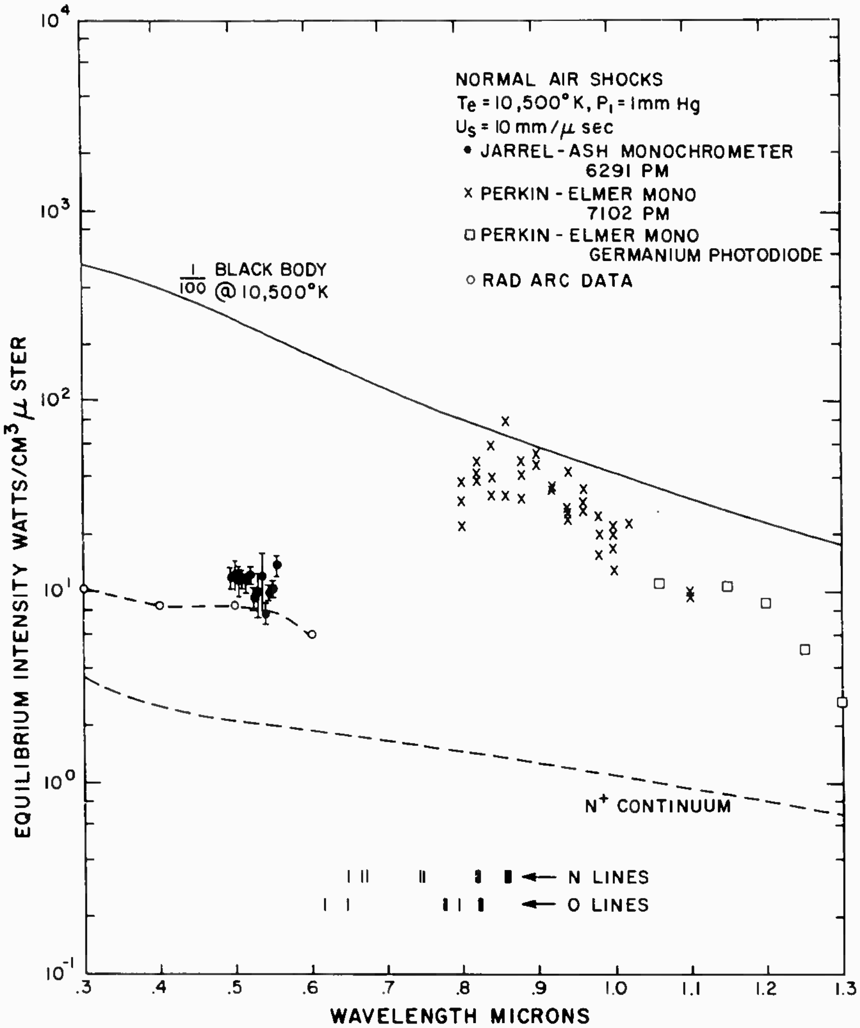

Fig. 5 Photometric measurements obtained in adjacent 50 Å channels between 5000 and 6000 Å of the equilibrium radiation behind 10 mm/μsec air shocks, $P_1 = 1.0$ mm Hg. The measurements show this wavelength region to be primarily a continuum. Also plotted in the figure are the arc measurements of Morris and Boch[12] performed at the same temperature in nitrogen and scaled by a small factor to the same product of neutral nitrogen atom and electron concentration

A1606

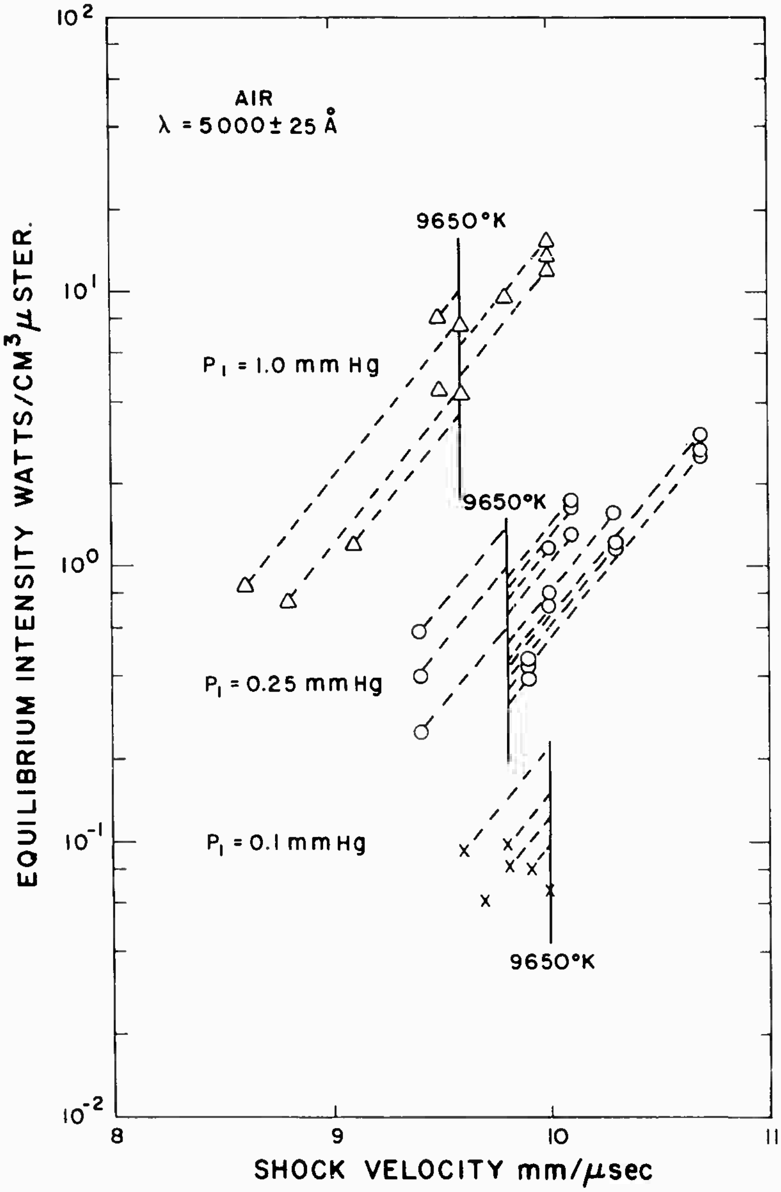

Fig. 6 Measurements obtained of the continuum radiation between 5000 and 5300 Å at three conditions of initial pressure. The measurements in the vicinity of 9650°K are extrapolated to their corresponding values at 9650°K.

A 1607

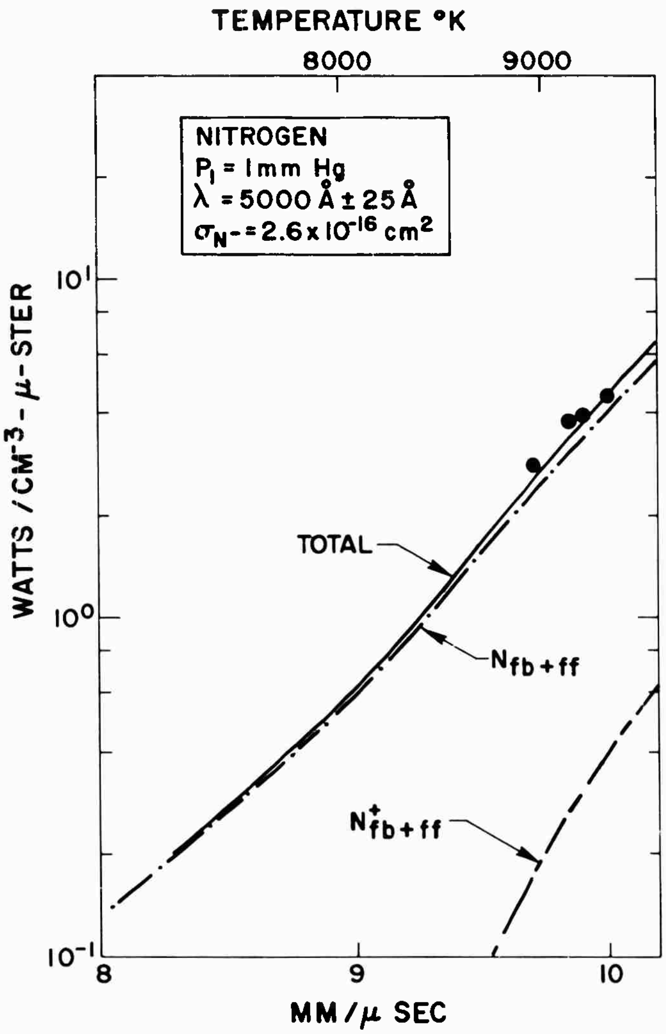

Fig. 7 Equilibrium measurements obtained from nitrogen shocks, P_1=1.0 mm Hg, the theory line is discussed in the text.

A1363

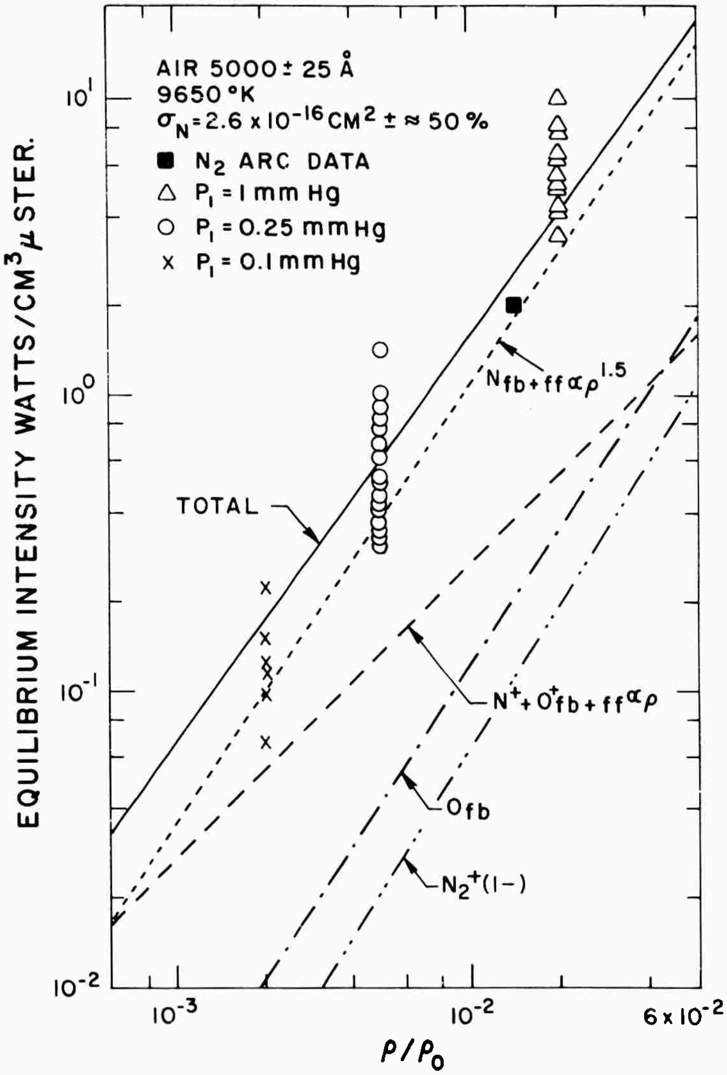

Fig. 8 The extrapolated photometric measurements of Fig. 6 to 9650°K plotted versus density. The lines are based on various theories discussed in the text. The strong density dependence of the radiation as well as the fact that the measurements fall above the calculated sum from other sources suggests the existence of an $N^-(^1D)$ continuum.

A1608

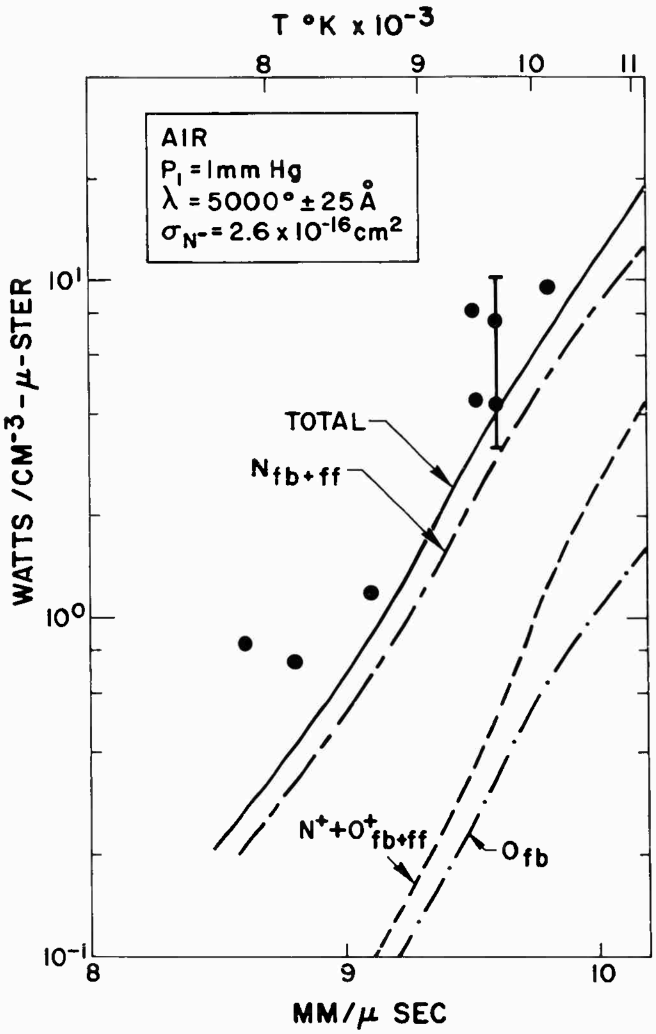

Fig. 9 Equilibrium measurements from incident shocks at 5000 Å in air, P_1=1 mm Hg, versus shock velocity. The radiation in this velocity range is deduced to be predominantly the $N^-(^1D)$ continuum radiation. The theory lines are discussed in the text.

A1364

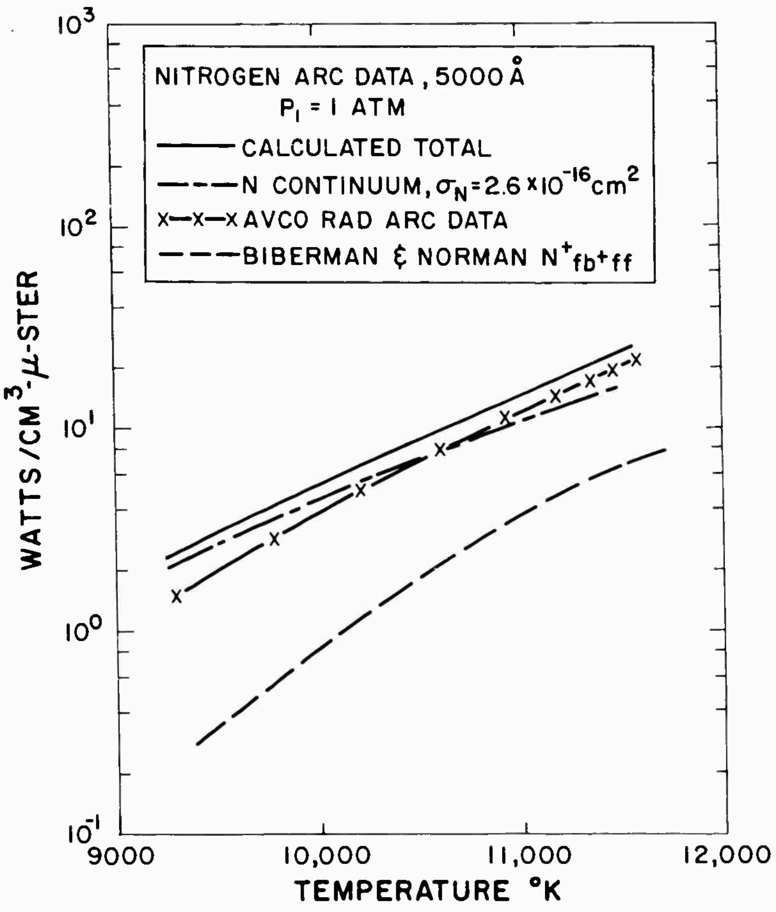

Fig. 10 Nitrogen arc data obtained by Morris and Boch[12] showing how the data are fit by assuming that the radiation is mainly that produced by the $N^-(^1D)$ continuum.

A1366

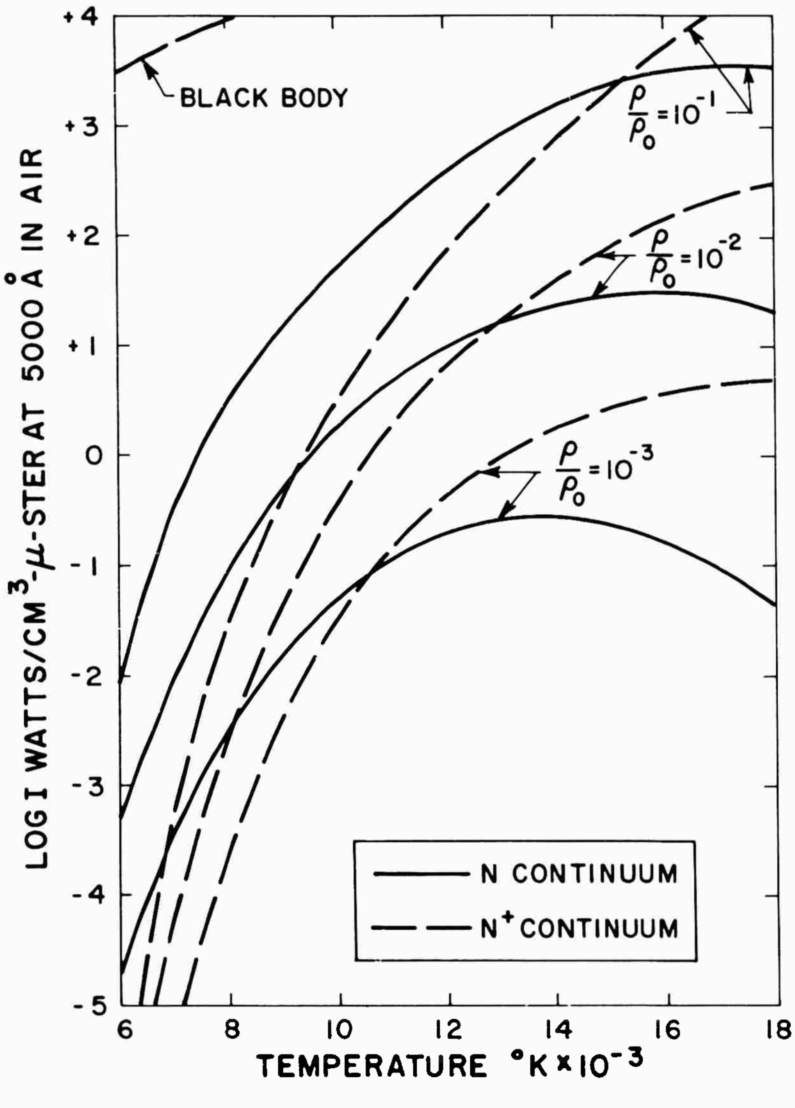

Fig. 11 A compairson between the $N^-(^1D)$ continuum and N^+ ion Kramers' radiation versus temperature at various densities. For super-satellite re-entries which decelerate at low altitudes, the $N^-(^1D)$ continuum could produce significant radiative heating.

A1368

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This work represents a study of the continuum radiation from shock-heated air and nitrogen as a function of density and temperature. For air or nitrogen in which the molecules are completely dissociated yet only partially ionized, radiation produced by the interaction of electrons with neutral atoms will have a stronger density dependence than the radiation produced by electron interaction with positive ions. Intensity measurements of the continuum at 5000 Å were made on the equilibrium region behind incident air and nitrogen shocks in the 9 to 11 mm/μ sec range at initial pressures of from 0.1 mm

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1. Air, shock-heated - Continuum radiation.
2. Nitrogen, shock-heated - Continuum radiation.
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VII. Contract NAS 9-858.

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